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Within the last two-and-a-half years, we have designed and characterized a few structures for generating and amplifying mid-IR waves based on GaAs/AlGaAs multilayers. We have implemented several new schemes for efficiently generating blue and green light. We have grown and characterized several structures towards optically-pumpled three- and four-level intersubband lasers. We have investigated a few new schemes for efficiently generating and amplifying THz to submm waves based on novel structures or schemes.

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Project Summary

Within the last two-and-a-half years, we have designed and characterized a few structures for generating and amplifying mid-IR waves based on GaAs/AlGaAs multilayers. We have implemented several new schemes for efficiently generating blue and green light. We have grown and characterized several structures towards optically-pumped three- and four-level intersubband lasers. We have investigated a few new schemes for efficiently generating and amplifying THz to submm waves based on novel structures or schemes.

1. Introduction

In February 1994, a research project entitled "Optoelectronic devices based on novel semiconductor structures" was initiated under the funding of AFOSR (Grant F49620-94-1-0154). The purpose of the project was to continue studying properties of semiconductor quantum well (QW) and superlattice (SL) structures for novel applications in nonlinear optical and optoelectronic devices. The project has been successfully carried out in collaboration with Prof. J. B. Khurgin. In May 1997 this project has been renewed for three more years (Grant F49620-97-1-0350). During the last two and a half years, a lot of exciting results have been obtained, as summarized below.

Multiple QWs and SLs have been the main focus of interest for many researchers since the sophisticated methods of material growth – molecular beam epitaxy and chemical vapor deposition had become mature, i.e. since late 70's. During 1980's, nonlinear optical and electro-optic properties of square QWs and SLs were investigated and shown to be different from the properties of bulk materials. Strong third-order nonlinear effects were predicted and later observed in QWs [1] and SLs [2] and attributed to band non-parabolicity [3,4], exciton line saturation [5], and quantum-confined Stark effect [6,7]. Practical devices using these effects, e.g. self-electro-optic effect devices (SEEDs) and electroabsorption modulators [8], have been successfully demonstrated.

The desire to improve the performance of nonlinear optical devices based on QWs and SLs has lead to the idea of using asymmetric heterostructures [9-14]. In asymmetric heterostructures symmetry is removed by either grading [15], strain [16], doping [17], or using asymmetric coupled or stepped QWs [18,19]. Asymmetric heterostructures were shown to possess large second-order susceptibility, and therefore, they could be used for frequency doubling, generation of tunable or multiple frequencies, optical rectification, and linear modulation of light. We have shown that switching and bistability [20] should be easier to achieve in asymmetric structures, since second-order susceptibility is larger than the third-order susceptibility.

Recently, surface-emitting green light was obtained [21] by frequency-doubling infrared laser beam (1.06 μm) in the waveguide based on periodically-modulated second-order susceptibility in alternating $Al_xGa_{1-x}As$ and $Al_yGa_{1-y}As$ ($x \neq y$) layers. When a thin AlGaAs layer is sandwiched between two quarter-wave stacks, large increase in the conversion efficiency was observed [22] due to cavity enhancement. Following Ref. [10], second-order susceptibility of

asymmetric quantum-well domain structures was measured in the surface-emitting geometry [23]. The maximum conversion efficiency so far is still less than 1%/W.

Recently, we showed [24] that the conversion efficiency for second-harmonic generation (SHG) can be greatly improved by including cavities, and, using quasi-phase-matched semiconductor structures. Most recently, we considered possibility of achieving optical parametric oscillation and amplification based on multilayer and asymmetric QW domain structures in the transverse-pumping geometry [25]. Furthermore, we proposed novel practical schemes for implementing a class of nonlinear optical devices by cascading second-order nonlinearities of semiconductor structures. They include optical frequency shifters [26], optical phase conjugation [27,28], waveguide coupling [29], self-phase modulation [30], and optical power limiting [27].

Recently, we have shown that second-order optical nonlinearities can also be used to generate THz waves based on various parametric processes [31,32]. We have used novel configurations combined with novel structures.

Recently, there have been quite a lot of efforts for fabricating self-assembled quantum dots [33,34]. Because of the three-dimensional quantum confinement in these dots, nonradiative relaxation times for the carriers between intersubbands due to phonon scattering are much longer. Therefore, it is possible to efficiently generate electromagnetic radiation following the transition between quantized energy states. Thus, we may be able to eventually achieve stimulated emission in the domain from far-IR to THz.

Chapter 2 is devoted to our results achieved within the last two-and-a-half years.

The optical parametric oscillators and amplifiers, optical frequency shifters, frequency doublers, and the other nonlinear optical devices based on the cascaded second-order nonlinearities, have potential applications in generation of blue light, generation and amplification of tunable mid-IR light, optical communication, counter measure, ultrafast detection, sensor protection, real-time holography, or optical lithography. All of these devices of the same kind can be integrated into an array on one wafer. THz generators and amplifiers can be eventually used for spectroscopy, communication, and bio-medical imaging. Intersubband lasers can be used for remote sensing, spectroscopy, and communication. We believe the proposed research will make dramatic impact on optoelectronics if it is successfully carried out.

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2. Results achieved within the last two-and-a-half years (May 1997 – Aug. 1999)

During the past two-and-a-half year period, we have made great achievements in four areas:

I. Efficient generation and amplification of tunable mid-IR waves

II. New schemes of efficiently generating blue-green light

III. Effort of optically-pumped 4-level intersubband laser

IV. Generation and amplification of coherent THz to submm waves

As a result of our research, within the covered period we have published 19 refereed journal papers, 5 conference proceedings papers, and made 34 conference presentations. Among these presentations, five of them are invited talks. In addition, we have submitted 8 journal papers for publication.

The detailed descriptions of our achievements in the four areas for the period are as follows:

2.1 Efficient generation and amplification of tunable mid-IR waves

2.1.1 Design and growth of new multilayer structure for generating 2.66 μm by mixing 1.55 μm with 980 nm

Several years ago, we proposed to use transversely-pumped counter-propagating optical parametric oscillators and amplifiers (TPCOPOs and TPCOPAs) to efficiently generate and amplify mid-IR beams [1]. The advantage of this novel configuration is the large tuning range achieved by changing the propagation direction of the pump [2]. Furthermore, the devices are miniature and ultrastable since they do not require any cavity to achieve oscillation. Recently, we have successfully grown a multilayer structure. It can be used to amplify the input beam at 1.58 μm and to generate a new beam at 3.23 μm if a pump at 1.064 μm is present [3]. As a first step, we have characterized this structure based on reflection-second-harmonic generation, as presented in the next section.

Due to rapid development of erbium-doped fiber and diode lasers, we have just designed and grown a new multilayer structure. This device can be used to amplify the input beam at 1.55 μm and to generate a new, 2.66-μm beam in the present of a pump beam at 980 nm. This structure can be eventually integrated with a vertical-cavity surface-emitting laser (VCSEL). As a result, the pump beam inside the multilayers can be provided via current injection in a VCSEL.

We are currently characterizing this structure. Our results will be presented at the 1999 OSA Ann. Meet.

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2.1.2 First observation of high-order quasi-phase-matched second-harmonic generation in GaAs/AlAs multilayers in reflection geometry

In this section, we report our first results on observation of high-order phase-matching peaks for second-harmonic generation in GaAs/AlAs multilayers in reflection geometry and confirmed quadratic dependence of the second-harmonic pulse energy on pump pulse energy for the first time.

GaAs and $Al_xGa_{1-x}As$ possess large magnitude of second-order nonlinear susceptibilities. By integrating with commercialized diode lasers, it is possible to eventually fabricate monolithic devices for efficiently generating coherent visible light, especially in the blue-green [4] and mid-IR [5] domains via parametric processes. However, because these materials have negligible birefringence, multilayers have to be used for quasi-phase-matching (QPM). There are two configurations for achieving QPM: surface-emitting [4,5] and reflection [6,7]. Reflected second-harmonic generation (SHG) in the $GaAs/Al_xGa_{1-x}As$ multilayer was initially studied in Ref. [6] and later in Ref. [7]. However, QPM peak was not clearly identified in Ref. [6] since the obtained spectrum is extremely broad. On the other hand, QPM peak was not directly measured in Ref. [7]. Upon the detailed examination of Refs. [6,7], we believe the pump wavelength is near the first-order QPM region in both cases. Here we report our results on detailed investigation of SHG in reflection geometry ("reflection-SHG") in the GaAs/AlAs multilayer. For the first time, we measured the spectra of the reflection-SHG and identified high-order QPM peaks. Moreover, we confirmed the quadratic dependence of the SH pulse energy on the pump pulse energy. 15 pairs of alternating GaAs/AlAs layers were grown by Molecular-Beam Epitaxy. The thicknesses of each GaAs and AlAs layers are 806 Å and 955 Å, respectively. They were chosen for achieving transversely-pumped counter-propagating optical parametric amplification

(TPCOPA) and difference-frequency generation (DFG) with a specific set of wavelengths [8]. A nanosecond laser pulse with tunable output wavelength was used in our measurement. We first measured the reflection-SHG spectra within 950-1220 nm at different incident angles ranging from 23° to 65°. In the measured wavelength range, three obvious peaks were found. Based on the QPM condition for the reflection-SHG, we have assigned two peaks to the QPM reflection-SHG with m = 2 and 3, respectively. We would like to stress that this is the first time to directly measure QPM reflection-SHG peaks in multilayers. The pump wavelength required to observe the first-order QPM peak is beyond the tuning range of our laser. On the other hand, certain peaks can be produced in the reflection direction for the SH beam by Distributed Bragg Reflection (DBR). The third peak is attributed to DBR at the first order. We also measured the dependence of the SH pulse energy on the pump energy per pulse. Our data exhibit a clear quadratic dependence. However, such a dependence was not confirmed previously in Refs. [6,7]. We have determined the conversion efficiency to be about 1.8×10^{-9}% at a peak pump intensity of 1.64 MW/cm^2. It is low because the generated SH beam is partially absorbed by the AlAs layers. We have re-designed the thicknesses and aluminum concentration of the layers to reduce absorption and to include a vertical cavity for the enhancement of conversion efficiency. We are in the process of measuring conversion efficiency on the optimized structure, and achieving TPCOPA and DFG. Besides the potential application for frequency-doubling, the reflection-SHG can be used to accurately determine indices of refraction above bandgaps of semiconductors for the first time as well as aluminum concentration and layer thickness.

As a result, we have submitted one paper to Opt. Commun. and presented our results at CLEO'99 and Quantum Optoelectronics'99.

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2.2 Novel schemes of efficiently generating blue-green light

2.2.1 First observation of backward quasi-phase-matched third-harmonic and second-harmonic generation

In this section, we summarize our results on observation of backward effective third-harmonic generation due to completely quasi-phase-matched cascaded second-harmonic and sum-frequency generation in periodically-poled potassium titanyl phosphate waveguide with domain period of 4 μm.

Recently, we achieved backward second-harmonic generation (SHG) in periodically-poled bulk $LiNbO_3$ (PPLN) [9]. We have improved the conversion efficiency by about an order of magnitude by using long laser pulses [10]. Here, we report our new results of backward SHG by using periodically-poled potassium titanyl phosphate (PPKTP) waveguide. More importantly, we report our results on first observation of backward third-harmonic generation (THG) due to *completely* quasi-phase-matched cascaded SHG and sum-frequency generation (SFG) in PPKTP waveguide. This is the first time to achieve backward effective THG where both of SHG and SFG are quasi-phase-matched. Previously, we observed backward THG in PPLN by cascading SHG and SFG [11]. However, only one of these two constituent processes is quasi-phase-matched. Therefore, the previous conversion efficiency is quite low. The hydrothermally-grown KTP substrate was poled by applying several 2.1 kV pulses for the duration of 400 μs with a spatial period of 4.0 μm. Channel waveguides (perpendicular to the domain-inverted grating) with dimensions of 4 μm × 6 μm were fabricated on the original +z face of the substrate by the standard ion-exchange process in a $RbNO_3$ / $Ba(NO_3)_2$ melt. The sample length is 2.6 mm. A nanosecond laser pulse with tunable output wavelength was focused and coupled into one of the waveguides by a microscope objective. We first measured backward SHG phase-matching spectrum. Five peaks at 1397.4 nm, 1328.0 nm, 1281.4 nm, 1231.7 nm, and 1181.3 nm were observed, which correspond to m_1 = 21, 22, 23, 24, and 25, respectively, for the backward SHG phase-matching condition

$$k_{2\omega} + 2k_{\omega} = 2\pi m_1 / \Lambda \tag{1}$$

where k_{ω}, $k_{2\omega}$ are wave vectors at pump and SH wavelengths, Λ = 4.0 μm is the spatial period of the domains and m_1 is an integer for QPM grating order. As a result, we achieved the conversion

efficiency of 0.6% for the pump energy per pulse of $J_P = 16$ μJ. Compared with 0.3% achieved in PPLN at much higher pump pulse energy (~ 300 μJ), this is a dramatic improvement. Fig. 6 shows the spectrum of backward THG. There are one *dominant* peak at 1233.7 nm (shown by the arrow) and seven secondary peaks at 1444.3 nm, 1404.6 nm, 1362.3 nm, 1323.4 nm, 1289.5 nm, 1258.1 nm, and 1227.7 nm, respectively. There are two mechanisms to generate backward TH beam. First, pump beam interacts with the forward SH beam, as in configuration (a). The seven secondary peaks were assigned to this configuration successfully. On the other hand, pump beam can interact with the backward SH beam, see configuration (b). For a short-period grating, it is possible to quasi-phase-match both the backward SHG and subsequent backward SFG [12]. In this case, the pump wavelength needs to satisfy both Eq. (1) and the following condition for backward SFG

$$k_{3\omega} - k_{2\omega} + k_{\omega} = 2\pi m_2 / \Lambda \qquad (2)$$

where $k_{3\omega}$ is the wavevector at the TH wavelength, m_2 is an integer for backward SFG grating order. For $m_1 = 24$ and $m_2 = 13$, both equations can be satisfied at the pump wavelength of 1230 nm, which is close to 1233.7 nm, determined from our experiment. Therefore, we believe that the dominant peak results from the cascaded backward SHG and backward SFG *with each second-order nonlinear process completely quasi-phase-matched.* The conversion efficiency is determined to be ~ 0.4% at $J_P \approx 35$ μJ. This is in good agreement with our theoretical value of ~ 0.99% based on Ref. [12]. There is a potential for us to improve the conversion efficiency to at least 10%.

As a result of our research, we have published two journal papers in Opt. Lett. and IEEE J. Quantum Electron. and presented one paper at CLEO'99. We will give an invited talk at Photonics West'00.

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2.2.2 Efficient generation of blue light by frequency-tripling single 1.32-μm beam in two separate KTP and Ce:KTP crystals

In this section, we report our results on efficient generation of blue light by frequency-doubling in undoped KTP and subsequent sum-frequency generation in Ce-doped KTP.

By frequency mixing of 1.064 μm and 0.809 μm radiation in a $KTiOPO_4$ (KTP) crystal, it is possible to generate blue light [13], however, in the presence of two pump beams. By cascading second-harmonic generation (SHG) and sum-frequency generation (SFG) in two separate $LiNbO_3$ and KTP crystals, coherent blue light can be generated by using a single pump beam from Nd:YAG laser (at e.g. 1.319 μm) [14]. However, this scheme has not been implemented solely in KTP crystals. KTP is one of a few important nonlinear optical materials because it is highly resistant to laser damage. In addition, it possesses large nonlinear optical coefficients with broad angular and temperature phase-match acceptance width-length products. However, it has noticeable absorption in the blue domain. Here, we report our results of reducing absorption of flux-grown KTP crystals in the blue domain by doping them with Cerium atoms (replacing ~ 4.8% of Titanium atoms). In the blue to green region the absorption coefficients of the Ce-doped KTP are significantly less than those of the undoped KTP. Therefore, we can achieve a relatively higher conversion efficiency in this domain by using a Ce:KTP crystal while minimizing the effect of laser heating. We have then used an undoped KTP crystal (5 mm × 5 mm × 5.7 mm, $\theta = 80.7°$, and $\phi = 0°$) to generate a coherent beam at 0.66 μm via SHG from a pump beam at 1.32 μm. The 1.32-μm pump beam is the output of a nanosecond optical parametric oscillator pumped by a frequency-tripled Nd:YAG laser, with the tunable output wavelength. By mixing the residual pump beam with the SH beam in a Ce-doped KTP crystal (5 mm × 5 mm × 8 mm, $\theta = 87°$, and $\phi = 0°$) via type-II SFG, we have successfully generated coherent blue light at 0.44 μm by using a *single* pump beam. In our measurement, the pump beam propagates along the yz plane in both crystals that were placed close to each other. A positive lens with a focal length of 10 cm was used to focus the pump beam into both crystals. For the SHG in the undoped KTP, the polarization of the pump beam forms 45° with the z axis and the generated SH beam is polarized normal to the z axis. For the SFG in the Ce-doped KTP, the 0.66-μm SH and residual 1.32-μm pump beams are polarized parallel to and normal to the z axis, respectively. The effective phase-matching wavelength for frequency tripling is 1.320 μm.

At this pump wavelength, we confirmed the cubic dependence. The overall conversion efficiency from the pump beam to the blue beam achieved so far is ~ 5.9% at a pump pulse energy of ~ 140 μJ. We would like to note that our peak pump intensity is much lower than those used before [13,14]. To further improve the conversion efficiency, we can use a longer SHG KTP crystal, a cavity, two separate optical set-ups for SHG and SFG, better beam profile of our pump laser, and/or higher pump energy per pulse. We can readily increase the overall conversion efficiency to about 20%. We will also use sub-picosecond laser pulses from our OPA system to achieve higher conversion efficiency. Our ultimate goal is to use the 1.319-μm output beam from our Nd:YAG laser to pump the undoped KTP and Ce:KTP crystals to commercialize an all-solid-state, compact, coherent blue light source.

We have submitted one journal paper to Opt. Lett., presented our results at CLEO'99, and given an invited talk at NOMA'99.

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2.2.3 Characterization of submicron-period ion-exchanged KTP waveguides based on backward second-harmonic generation

Periodically-segmented ion-exchanged $KTiOPO_4$ (KTP) waveguides can be used for not only quasi-phase-matched (QPM) second-harmonic generation (SHG) [15], but also acting as a distributed Bragg reflector (DBR) due to index grating [16] to enhance conversion efficiency for SHG. In this case, a submicron-period index grating is desirable because it can reach a given reflectivity in a short length. One way to characterize the waveguide containing a submicron-period index grating is to measure reflection spectrum of an incident beam. Due to the presence of the index grating, it is also possible to use SHG to characterize the grating. However, in such a short-period grating, the forward SHG can *never* be phase-matched. Therefore, backward SHG [17] can be used as a novel and unique technique to characterize a short-period grating. This technique has an advantage for determining interface roughness, fluctuation of index layer thickness from one period to the next, and duty cycle. In addition, one can use this technique to

determine whether the domain is inverted after ion exchange. Previously, it was demonstrated that domain inversion can still occur for the period as short as 3 μm [18]. Here, we report our results of achieving QPM backward SHG in periodically-segmented ion-exchanged KTP waveguide with the period of 0.7 μm. We have fabricated 4-μm-wide periodically-segmented KTP waveguides on a z-cut flux-grown KTP substrate by ion-exchange in a $RbNO_3/Ba(NO_3)_2$ melt at 350°C for 45 minutes. The diffused segments were 0.4-μm-long RTP separated by 0.3-μm-long KTP. There are 3200 periods in the center of the waveguide, which corresponds to a total length of 2.24 mm for the segment portion. A mode-locked Ti:sapphire Laser with a pulse width of ~130 fs and repetition rate of 76 MHz was used as the pump source. The polarization of the fundamental beam is always parallel to the z-axis of the crystal for utilizing d_{33}. We first measured the average power of the backward SH beam while scanning the fundamental wavelength from 750 to 910 nm with a constant average pump power across the spectrum. For each center pump wavelength, we measured the SH spectrum. We then determined the peak power and corresponding SH wavelength. By varying the center pump wavelength, we obtained a spectrum for backward SH beam. There are four large peaks at 384.5, 395.6, 440.7 and 450.3 nm. We have identified two peaks to the 7^{th} (385.8 nm) and 6^{th} (442.7 nm) backward QPM SH peaks, and the other two to the 7^{th} (396.9 nm) and 6^{th} (452.2 nm) DBRs, respectively. The dependence of the average backward SH power on the average pump power exhibits an excellent quadratic characteristic. At the input power of 125 mW, the average output SH power is about 200 nW. The conversion efficiency is $\sim 1.6\times 10^{-4}$%. Our theory predicted the conversion efficiency to be $\sim 5\times 10^{-4}$% following Ref. [19]. This is in good agreement with our experimental result. Based on our results and measurements, we believe our RTP domains are *inverted* as a result of ion exchange. This breaks the previous record of the domain inversion for the shortest period of 3 μm. The measured linewidth is ~ 0.7 nm, which is much larger than our estimated value of ~ 0.023 nm. Following Ref. [20], we have determined the variance in the period to be 4.5×10^{-3} μm. Since backward SHG has a much narrower linewidth for an ideal domain structure, it can be eventually developed to a novel and sensitive technique for measuring the fabrication errors of the gratings.

We have submitted our results to Appl. Phys. Lett. for publication and presented some of our results at OSA Ann. Meet.'98. We presented our updated results at CLEO'99.

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2.2.4 Investigation of damage threshold of $KTiOPO_4$ crystals using CW Argon laser towards intracavity second-harmonic generation

Recent development of lasers and nonlinear optical materials made it possible to commercialize CW high-power all-solid-state laser-diode (LD) pumped green lasers such as Millennia and Verdi. The nonlinear material used so far is limited to LiB_3O_5 (LBO). Its disadvantage is relatively high temperature required for achieving second-harmonic generation. On the other hand, potassium titanyl phosphate ($KTiOPO_4$ or KTP) possesses a large second-order susceptibility, and large angular and temperature bandwidths for frequency doubling 1.064-μm source. Most importantly, it is possible to generate green light from 1.064-μm pump beam at room temperature. High-power LD pumped green lasers using KTP have been already implemented with the CW output power as high as 6.3 W [21]. The maximum output power that can be achieved in KTP depends on its damage threshold. There are quite some results on reporting different damage thresholds, however, only using pulsed lasers [22]. Thus, it is important for us to develop a routine method to evaluate KTP crystals towards CW frequency-doubling applications.

In this report, we present a simple method for assessing KTP crystals. We have used a CW Argon laser as a pump source with its output wavelengths primarily consisting of 488 nm and 514 nm. The pump beam is focused onto a KTP crystal via a converging lens. The beam size at the crystal is about 23 μm. We have studied eight pieces of KTP samples with different sizes. While most of them are undoped others are doped with Rubidium and/or some of them are anti-reflection-coated. These samples were cut from a large size single crystal grown by using high-temperature solvent (flux) method in which the raw high-purity TiO_2, KH_2PO_4, K_2CO_3 materials

were used with some dopants and the solvent $K_6P_4O_{13}$. The samples were correctly oriented for type-II phase-matching at 1.064 μm at room temperature. At relatively low laser powers (larger than 0.2 Watts), there is only invisible damage regardless of polarization. It can be seen in the transmission patterns formed after passing the Argon beam through the crystal while moving the beam along z axis. The damage area always occurs next to the beam. At relatively higher laser powers (0.2-0.5 Watts) and with the beam polarized normal to z axis, there is black coloration (darkening of crystals or gray tracks) of a crescent shape formed. In undoped KTP crystals, for every dark line formed, there is always an orange dot formed above the dark line. However, when the polarization of the pump beam is parallel to the z axis, there is no black coloration observed for the laser power as high as 4.5 Watts. Following Ref. [23], due to two-photon absorption and third-harmonic generation, electrons and holes can be generated. Most of these electrons and holes will quickly recombine. However, some of them will be trapped at stabilizing defect sites. These defects trapped by the electrons and holes produce additional absorption around 500 nm. As a result, a significant portion of the laser beam is absorbed. The absorption of the laser beam causes heating of local area. Because KTP is a pyroelectric crystal [24], a dc electric field can be generated in the laser-beam area upon heating. Under such a dc electric field, potassium ions can move towards -z axis, see Ref. [25]. We believe the non-uniformly accumulated potassium ions next to the beam are responsible for the observed transmission patterns. Following Ref. [26], there is a more intense optical absorption which has a lifetime of 10 ms at room temperature. It is probably due to holes trapped at a bridging oxygen ion between two titanium ions and stabilized by a nearest-neighbor potassium vacancy, and electrons trapped at perturbed Ti sites (Ti^{3+}). Although this absorption is short lived, it may play the most important role in the formation of black coloration. Since we used a CW laser beam, the trapped holes do not decay within 10 ms. As a result, the absorption can be enhanced due to accumulation of trapped electrons and holes within 10 ms. Such intense absorption causes local heating, and therefore induces a large dc electric field through pyroelectric effect. More and more potassium ions drift out of the laser-beam area. In addition, more holes and electrons will be trapped near potassium vacancies and perturbed Ti sites. Eventually, there will be a local region of the trapped electrons and holes, which forms gray tracks. They preferentially absorb light whose electric vector (i.e. polarization) lies along the c axis (normal to z axis), see Ref. [27]. According to Ref. [28], the preferred absorption is mainly due to electrons trapped at

perturbed Ti sites (Ti^{3+}). Since gray tracks preferentially absorb light with its polarization along c axis (normal to z axis), when the polarization of the laser beam is normal to z axis, the absorption is the strongest. The generated dc electric field is the strongest. The damage threshold is the lowest. On the other hand, the holes can also be trapped into the impurities such as Fe^{3+}. The orange dots may be due to the presence of these impurities trapped by holes.

For the first time, we have observed photorefractive effect in KTP crystals. For two pump beams with a power of 100 mW each and an angle of 60° between them, the diffracted power is ~ 0.01 mW in both polarization states.

We will further investigate the damage mechanisms. Based on our investigation so far, we are confident that KTPs grown by us can be used to generate CW coherent green light via frequency doubling 1.064-μm pump beam with the output power as high as 2 Watts. In the future, we plan to measure damage thresholds of LBO grown by us such that we can compare the results based on KTP and LBO. We also plan to commercialize a system similar to Millennia or Verdi for producing CW 2-W green light based on KTP crystals.

As a result of our study, we have submitted our results to Opt. Lett., published our results in the SPIE Proceedings, and presented our results at Photonics West'99 and OSA Ann. Meet.'98. We will present our updated results at the OSA Ann. Meet.'99.

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2.3 Effort of optically-pumped 4-level intersubband laser

2.3.1 Anomalously large blue shift of donor-acceptor pair transition peak in GaAs/AlGaAs coupled quantum wells

As excitation laser intensity increases, due to the change of Coulomb interaction energy between the recombined donors and acceptors, the donor-acceptor pair (DAP) transition energy shifts to high energy (i.e., blue shift) [29]. Usually this blue shift is very small (<3 meV) in bulk semiconductor materials with low impurity density [29,30]. Previously, we reported a large blue shift of 11 meV in GaAs/AlGaAs compensation-doped quantum wells (QWs) with the doping density of 3×10^{17} cm^{-3} [31]. However, at the relatively high laser intensities, the free-excitonic emission dominates the photoluminescence (PL) spectrum with the DAP transition as a shoulder. The peak energy of the DAP transition was obtained only after the PL profile was decomposed into two peaks. As a result, we did not directly observe the large amount of the blue shift.

Here, we report our results on direct observation of an anomalously large blue shift as large as 12 meV in GaAs/AlGaAs QWs with low residual (undoped) impurity densities. Each PL spectrum consists of only the DAP transition peak within the entire range of pump intensities. Therefore, the amounts of blue shift determined here are much more accurate than those in Ref. [31]. The sample was grown by molecular-beam epitaxy on a [100] semi-insulating n-type GaAs substrate. The epitaxial layers consist of 10 periods, each of which is composed of two GaAs QWs with widths of 50 and 65 Å, respectively. The two wells were coupled by a 100-Å $Al_{0.25}Ga_{0.75}As$ barrier. The adjacent periods were isolated from each other by 300-Å $Al_{0.25}Ga_{0.75}As$ barriers. All the layers were grown at the temperature of 590 °C with an average growth rate of 1.0 μm/hr for GaAs layers and 1.33 μm/hr for the $Al_{0.25}Ga_{0.75}As$ layers. The residual donors and acceptors in QWs are Si and C, respectively. We measured PL excitation spectrum at 4 K and the energy of the PL emission in the $Al_{0.25}Ga_{0.75}As$ layers and thus confirmed the aluminum concentration. As the excitation intensity increases from 43.6 W/cm^2 up to 1396 W/cm^2, the energy of the DAP transition peak shifts towards the high energy side (blue shift). The maximum shift is about 12 meV. To the best of our knowledge, this is the largest blue shift of the DAP transition peak ever observed in the unintentionally-doped QWs. Due to the imperfections at the interfaces between GaAs and $Al_{0.25}Ga_{0.75}As$, the residual donors and acceptors are most likely trapped at the interfaces. Under such an assumption, even low three-dimensional impurity densities can lead to a large area density at each interface. Based on this

model, the shift of ~12 meV is due to the change of the Coulomb interaction energy when the separation between the recombined donors and acceptors decreases to $R_0 \approx 92$ Å, (or a local interface area impurity density of 3.8×10^{11} cm^{-2}). Since the width of the wide QW in each unit is 65 Å, one should also observe the PL emission peak due to the recombination between the donors at one interface and acceptors at the other one. When the sample temperature is increased to 40 K, the PL peak wavelength is at 791 nm. The energy of this peak is larger than the lowest energy of the DAP transition peak at 4 K by 17.6 meV. Therefore, we have assigned this peak to the recombination between the donors at one interface and acceptors at the other one. We believe this is the first observation of such a type of the DAP transition.

By characterizing the DAP transition peaks, we can determine doping locations and profiles, which are essential for achieving intersubband lasers.

Our publications include a journal paper submitted to Appl. Phys. Lett., a presentation at QELS'99, and an invited talk at Quantum Optoelectronics'99.

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2.3.2 Observation of saturation of excitonic transition peak in GaAs thin film at anomalously low laser intensities

In this section, we report our observation of obvious saturation of excitonic transition peak in bulk-like GaAs when increasing laser intensity, due to exciton linewidth broadening as a result of scattering of excitons by other excitons and carriers.

In semiconductor bulk or quantum wells [32] excitonic transition peak can be eventually saturated by increasing laser intensity. However, the saturation occurs at high pump laser intensities of 0.58 kW/cm^2 for quantum wells and much higher for bulk GaAs. Recently, we observed saturation of excitonic emission peak in quantum wells after growth interruption at every interface at extremely low laser intensities [33,34]. Here, we present our new results of photoluminescence (PL) and photoluminescence excitation (PLE) studies of a GaAs thin film.

For the first time, we have observed the saturation of the excitonic transition peak at extremely low laser intensities. We believe it is due to linewidth broadening of the exciton peak as a result of scattering of excitons by other excitons and carriers. The undoped GaAs thin film of thickness of 1 μm was grown on a [100] semi-insulating n-type GaAs substrate at the growth rate of 1.0 μm/hour and growth temperature of 590 °C by molecular-beam epitaxy. The residual donors and acceptors in the thin film are Si and C, respectively, with their average densities in the range of 10^{14}-10^{15} cm^{-3}. We first measured PL spectra at the temperature of 4.1 K and the pump wavelength of 790 nm. The observed four peaks with the corresponding wavelengths of 817.9, 819.0, 822.3 and 831.2 nm correspond to free-exciton (X), donor-bound exciton (DX), acceptor-bound exciton (AX), and conduction-band-to-acceptor (CA) emission peaks, respectively [35]. When laser intensity is increased from 8.7 W/cm^2 to 436 W/cm^2, the emission peaks, CA, AX and DX, are gradually saturated, due to partial filling of the impurity sites. We then measured the PLE spectra at different laser intensities. We have assigned the sharp transition peak to the free-excitonic absorption peak and the broad one to the superposition of the DX and AX absorption peaks. As the laser intensity increases, the relative strength of the free-exciton peak decreases. At the pump intensity of 524 W/cm^2, it completely disappears. In addition, the linewidth (HWHM) increases from 0.79 meV to 1.8 meV when the intensity increases from 0.087 W/cm^2 to 524 W/cm^2. This is the first observation of such a large increase of the free-exciton linewidth at such low laser intensities. To the best of our knowledge, 0.79 meV is the narrowest linewidth ever achieved in thin films [35]. Because of such a narrow linewidth, the lifetime of excitons can be readily decreased due to the scattering of the excitons participating radiative recombination by other excitons and electrons. As the exciton lifetime is decreased, the exciton linewidth is thus increased. Previously, exciton-exciton and exciton-electron scattering was observed in bulk-like GaAs, however, at much larger laser intensities [36]. We are currently measuring of exciton lifetimes to confirm the proposed mechanism for the linewidth broadening, and therefore, the saturation of the free-exciton transition peak.

These results can be used to determine the densities of the residual impurities of the grown structures.

We are currently preparing a journal paper for publication.

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2.4 Generation and amplification of coherent THz-submm waves

2.4.1 Efficient generation and amplification of tunable THz to submm waves based on parametric processes

In this section, we consider to use CdSe, GaSe, periodically-poled $LiNbO_3$, $LiTaO_3$, and diffusion-bonded-stacked GaAs and GaP plates for efficient generation and amplification of tunable submillimeter to THz waves, based on forward and backward parametric processes.

Second-order optical nonlinearities can be used to generate incoherent terahertz waves based on parametric fluorescence [37] and coherent ones via difference-frequency generation (DFG) [38]. To generate THz waves based on optical parametric oscillation (OPO) one needs a nonlinear crystal that has high damage threshold and spatial homogeneity. Recently THz–OPO was implemented in bulk $LiNbO_3$ [39]. The threshold for OPO can be reduced by cooling $LiNbO_3$ to liquid-nitrogen temperature [40]. The advantage of using birefringence is tunability of THz wavelength through crystal rotation. However, there are some inherent disadvantages of using $LiNbO_3$ crystals. First of all, in the THz domain, the absorption coefficients of $LiNbO_3$ are large due to presence of the A_1–symmetry polariton mode. Secondly, the birefringence is too large for collinear phase-matching. As a result, in Refs. [39,40], a noncollinear propagation configuration was used. In addition, a short propagation length for THz was used and grating was used to effectively couple THz waves out of the $LiNbO_3$ crystal. However, because of this noncollinear propagation configuration, effective interaction length among the pump, signal, and idler waves is short for a small focused beam size of the pump. In Refs. [39,40], however, the

interaction length is limited by the effective absorption length.

In order to improve conversion efficiency, one needs to use parallel configurations such as the forward and backward ones. More importantly, one should look into possibilities of using other nonlinear optical materials that have lower absorption coefficients and appropriate amount of birefringence for phase-matching. Only very recently, quasi-phase-matching (QPM) became a practical alternative to birefringence-based phase-matching, for achieving coherent radiation covering the range from UV to mid-infrared [41]. Recently, we proposed to use periodically-poled $LiNbO_3$ (PPLN) to achieve THz waves based on the forward or backward OPO, or DFG [42]. In the presence of a single pump beam with its power below the OPO threshold, we can generate parametric fluorescence for the forward configuration without a cavity [42]. However, it is feasible to generate THz waves from a single pump beam for the backward configuration without any cavity [42]. Other materials such as $LiTaO_3$ or $KTiOPO_4$ can also be poled for efficiently generating THz waves.

It is obvious that the magnitude of second-order susceptibilities is one of the most important parameters for efficiently generating THz waves. Among different classes of second-order nonlinear optical materials, semiconductors such as GaAs, GaP, ZnSe, GaN, etc. possess large second-order susceptibilities. However, most of these materials are cubic crystals. Thus, the birefringence effect is too small for phase matching. Recently, an alternatively-rotated stack of GaAs plates has been successfully fabricated based on diffusion-bonding method [42]. Via this technique, dipole moments or second-order susceptibilities can have opposite signs between any two adjacent plates. As a result, QPM has been achieved for efficiently generating mid-infrared waves [43]. Obviously, other materials such as GaP can also be fabricated into a stack of the plates for QPM. By properly designing the thickness of each plate, these stacks can be also used for generating THz waves. The disadvantage of using the stacks is the unavoidable scattering loss at the interfaces and the limited number of the GaAs or GaP plates that can be diffusion-bonded [43].

Besides using the parametric processes for THz generation, there are also other competing approaches. One of them that have attracted much attention is based on broadband THz radiation from charge transport and optical rectification [44]. However, the disadvantage of generating THz waves based on this scheme is temporal-incoherence of the THz waves. As a result, one cannot study narrow absorption lines with these THz waves.

We have considered CdSe, GaSe, periodically-poled $LiNbO_3$ and $LiTaO_3$, and diffusion-bonded-stacked GaAs and GaP plates for OPO, OPA, and DFG [45]. CdSe is a positive uniaxial crystal ($n_e > n_o$). It has a transparency range of 0.75–25 μm. For efficient generation of the mm–THz waves, the absorption coefficients in the mm–THz domain are very important. However, the absorption coefficients measured so far on CdSe are limited to the wavelength of 72 μm. Based on those available and the absorption coefficients for $LiNbO_3$, we have concluded that the absorption coefficients for CdSe in the mm–THz domain are less than those for $LiNbO_3$. As shown in Ref. [42], for the crystal thickness of 5 cm and the signal wavelengths longer than 250 μm, the absorption coefficients only increase the threshold pump intensities by factors less than five. The damage threshold reported is about 6×10^9 W/cm^2 for 100-ps laser pulses at 3-Hz repetition rate and wavelength of 2.8 μm [46].

We now consider the forward OPO. We have assumed that the pump wave is an extraordinary wave while the signal and idler waves are ordinary ones. Only for the short pump wavelengths the signal wavelengths may be within the absorption range of CdSe. We now choose a pump wavelength to be 10.6 μm. When the angle is tuned from 35° to 90°, the signal wavelength can be tuned from 993 μm to 333 μm. However, when the angle changes, the threshold pump intensity also changes. With L = 5 cm and $R_p = R_i = 99\%$, the lowest value is 8.8×10^5 W/cm^2 for $\theta = 90°$ ($\lambda_s = 333$ μm).

We then consider the backward OPO, i.e. "oeo" polarization interaction. Compared with the forward configuration, the signal can be generated in the THz–mm region for much shorter pump wavelengths. This is an advantage of the backward configuration since most commercialized lasers have powerful output radiation in the visible-to-near-infrared domain. When tuning the angle θ, one can tune the signal wavelength. When θ changes from 35° to 90° via rotation of the crystal the signal wavelength can be tuned from 956 μm to 313 μm. To consider the threshold intensity for the oscillation, we assume that the input and output facets are anti-reflection-coated at the pump and the idler wavelengths, but not at the THz wavelength. The minimum value is $I_{p,th} = 4.7\times10^8$ W/cm^2 for $\theta = 90°$ ($\lambda_s = 313$ μm), which is much below the damage threshold for CdSe [46].

The threshold intensity can be reduced by including a cavity for the pump and idler waves for the backward OPO. For example if $R_s = R_i = 99\%$ at 1.064 μm and 1.068 μm, respectively,

the threshold pump intensity is reduced to 4.4×10^4 W/cm^2 for $\theta = 90°$ ($\lambda_s = 313$ μm). If $I_p = 2 I_{p,th}$, $\eta = 50\%$.

We now discuss possibility of using a CdSe crystal to amplify THz waves. We assume that the input and output facets are anti-reflection-coated at the pump and idler wavelengths, but not at the signal wavelength. If $I_{in}/I_{p,th} \leq 3.4\times10^{-3}$, the dependence of the amplification factor (G) on the input intensity exhibits a power law with an exponent index of – 0.5. However, when $I_{in}/I_{p,th} > 3.4\times10^{-3}$, G is close to unity due to the severe saturation by the input intensity.

When the pump intensity is below the threshold for the oscillation, one can use backward DFG in the presence of the pump and idler waves to achieve THz output. As shown in Ref. [45], one can calculate the intensity of the THz (signal) wave. We again assume that the input and output facets are anti-reflection-coated at 1.064 μm and 1.068 μm, respectively. If the pump intensity is $0.01\times I_{p,th} = 4.7\times10^6$ W/cm^2, the signal intensity is 295 W/cm^2 at $\lambda_s = 313$ μm in the presence of the pump and idler waves at the respective wavelengths of 1.064 μm and 1.068 μm. If the pump and idler beams have their beam size of ~ 313 μm, the peak pump power is about 3.6 kW for the signal output power of ~ 0.23 W. This corresponds to a conversion efficiency of $6.2\times10^{-3}\%$.

Other birefringence-based materials can also be used to efficiently generate mm–THz waves. GaSe is a promising candidate. This negative uniaxial crystal ($n_o > n_e$) has a large transparency range of 0.62–20 μm and second-order susceptibility (53 pm/V). Furthermore, its absorption coefficients are around one order of magnitude lower than those for $LiNbO_3$ at the low-frequency end and several orders of magnitude lower at the high-frequency end. Compared with the absorption coefficients of CdSe, GaSe probably has lower values. Therefore, it is possible to use GaSe for efficient generation of the mm–THz waves at the frequency up to several THz.

Based on Ref. [47], assuming $\lambda_p = 2.8$ μm, $\lambda_s = 400$ μm, and "eoo" parametric interaction configuration, we can obtain $\theta = 18.5°$. If $\phi = 30°$ and $L \approx 1.2$ cm, we have estimated $I_{p,th} \approx 10^{10}$ W/cm^2. This value is certainly below the damage threshold of 30 GW/cm^2 reported in Ref. [48].

We now consider PPLNs. As shown in Ref. [42], if the pump wavelength is 0.532 μm, to achieve the signal wavelength in the range of 125–1000 μm based on the backward configuration one needs the spatial period of the PPLN domains in the range of 15.7–133 μm. The PPLNs with

these periods are commercially available. If one includes absorption coefficients into consideration, the minimum threshold intensity for the backward OPO is 8.2×10^8 W/cm^2 at the signal wavelength of 500 μm with the total thickness of the PPLN 5 cm. On the other hand, based on backward DFG, one can generate the signal intensity of 1.7×10^4 W/cm^2 at $\lambda_s = 500$ μm, for L = 5 cm and the pump and idler intensities of ~ 10^8 W/cm^2. In the presence of only a pump beam below the threshold for the OPO, one can only generate backward parametric fluorescence [42].

Since periodically-poled $LiTaO_3$ has most properties similar to PPLN, we would expect that the results for periodically-poled $LiTaO_3$ are similar to those for PPLN. However, the absorption coefficients in $LiTaO_3$ in the millimeter–THz domain can be two orders of magnitude larger than those for $LiNbO_3$. For example, at $\lambda_s = 500$ μm the absorption coefficients for $LiNbO_3$ and $LiTaO_3$ are 0.5 cm^{-1} and 49 cm^{-1}, respectively. Besides, the effective nonlinear coefficient for $LiTaO_3$ is about a factor of 2 smaller than that for $LiNbO_3$. These two major differences between $LiNbO_3$ and $LiTaO_3$ make $LiTaO_3$ much less attractive for efficient generation of mm–THz waves. However, one can still use periodically-poled $LiTaO_3$ with a small thickness to generate mm–THz waves via backward DFG. Since the refractive index at the THz wavelength does not change much within 100–1000 μm, the spatial period Λ is linearly proportional to the THz wavelength. For $LiTaO_3$ in the range of the signal wavelengths 125–1000 μm, the QPM period is in the range of 13.7–110 μm.

We now consider alternatively-rotated diffusion-bonded-stacked GaAs. If every other [100] GaAs plate is rotated by an angle of 180° first around x axis and then around y axis, effective nonlinear coefficients change their sign from one plate to the next.

We assume the backward propagation configuration in which the pump and idler waves propagate along the GaAs [$\bar{1}$10] direction with the same polarization that forms an angle of 35.3° with the GaAs [110] direction, i.e. parallel to the [111] direction. The polarization of the THz wave is also parallel to the GaAs [111] direction. In this case, the effective nonlinear coefficient is $d_{eff} = 61$ pm/V following Ref. [43]. For generating the mm – THz waves in the range of 100–1000 μm, the required plate thickness is within 7.2–72 μm. For $\lambda_s = 300$ μm and $\lambda_p \approx 2.8$ μm, one obtains $\Lambda \approx 43.3$ μm, i.e. each plate has a thickness of 21.7 μm. The thickness can be also a multiple of this value for QPM at a high order. Assuming that there are 100 diffusion-bonded

plates, the total thickness is about 2.2 mm. The beam propagation length is then 3.1 mm. We have estimated the threshold pump intensity to be 4.9×10^{10} W/cm^2. According to Ref. [43], this value is probably above the surface-damage threshold of the diffusion-bonded-stacked GaAs plates. If the pump intensity is much below the threshold for the backward OPO, we can use the pump and idler beams to achieve THz waves via backward DFG. Assuming $I_p = I_i \approx 10^9$ W/cm^2, one can estimate the THz (signal) intensity to be $I_s = 3.5\times10^5$ W/cm^2 ($\lambda_s \approx 300$ μm). For the beam diameter of 300 μm, this THz intensity corresponds to a peak power of about 246 Watts.

Since GaP has some properties similar to GaAs, it is possible to create a stack of alternatively-rotated diffusion-bonded GaP plates for the QPM generation of THz waves. In the mm – THz range, the absorption coefficients for GaP are larger than those for GaAs. For example, based on the data available for GaAs at $\lambda_s = 333$ μm the absorption coefficient is 0.42 cm^{-1} while for GaP at $\lambda_s \approx 366$ μm, the absorption coefficient is 1.3 cm^{-1}. In addition, for the same polarization and propagation direction as in Ref. [43] the effective nonlinear coefficient for GaP is 27 pm/V ($d_{14} = 37$ pm/V).

We can estimate the threshold pump intensity. For $\lambda_p \approx 2.8$ μm, $\lambda_s = 300$ μm (the plate thickness of 24 μm), and 100 plates ($L \approx 3.3$ mm), $I_{p,th} \approx 1.7\times10^{11}$ W/cm^2 for achieving the backward OPO. If the pump and idler input intensities are $I_p \approx I_i = 10^9$ W/cm^2 one can estimate the output intensity for the THz wave to be 1.0×10^5 W/cm^2 following Ref. [45]. For a beam diameter of ~ 300 μm, this translates into an output power of ~ 72 W for the peak pump and idler powers of 7.1×10^5 W.

We now consider the forward configuration in which all three waves propagate in the same direction. For GaAs, $\lambda_s = 300$ μm, 6 plates with each thickness of 602 μm, and $R_p = R_i \approx 0.95$, $I_{p,th} \approx 9.8\times10^7$ W/cm^2. At such an intensity, the pump intensity inside the cavity is below the damage threshold. For GaP, $\lambda_s \approx 296$ μm, $t_p \approx 516$ μm, 6 plates, and $R_p = R_i \approx 0.95$, $I_{p,th} \approx 5.1\times10^8$ W/cm^2. Such a pump intensity results in the pump intensity inside the cavity, that is probably still below the damage threshold for GaP.

Our publications include a journal paper to be published in Opt. Quantum Electron., a SPIE Proceedings paper, and two conference presentations at International Photonics Conference'98 and Photonics West'99.

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8 Pending Publications

X. Mu, Y. Wang, V. Petrov, I. B. Zotova, Y. J. Ding, and W. P. Risk, "First characterization of periodically-segmented submicron-period ion-exchanged $KTiOPO_4$ waveguide based on quasi-phase-matched backward second-harmonic generation," submitted to Appl. Phys. Lett.

Y. J. Ding, "Parametric amplification of short pulses in transverse-pumping geometry," submitted to JOSAB.

X. Mu and Y. J. Ding, "Direct measurement of anomalously large blue shift of donor-acceptor pair transition peak in undoped asymmetric-coupled quantum wells," submitted to Appl. Phys. Lett.

X. Gu, Y. J. Ding, W. S. Rabinovich, and D. S. Katzer, "Observation of reflection-second-harmonic generation in GaAs/AlAs multilayers," submitted to Opt. Commun.

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X. Gu, X. Mu, and Y. J. Ding, "Efficient generation of blue light in two separate KTP and Ce:KTP crystals," submitted to Opt. Lett.

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X. Mu, X. Gu, and Y. J. Ding, "Frequency-tripling of 1.32-μm beam in single Ce:KTP using subpicosecond laser pulses," submitted to Opt. Lett.